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Preparation of reaction-bonded silicon carbide with well controlled structure by tape casting method

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Abstract

Tape casting is a reliable and cost effective method for producing thin ceramic sheets with uniform and tailored microstructures, especially for multilayered composite materials. In this paper, SiC/C tapes were prepared by tape casting method. After lamination and binder removal, porous preforms with homogeneous microstructure and narrow pore sizes distribution were developed. Then, dense reaction bonded SiC ceramics (RBSCs) were obtained by silicon infiltration into these preforms. The highest bending strength of the RBSCs can reach 410 ± 14 MPa. Moreover, impregnation of phenolic resin into the porous preforms before silicon infiltration could help to develop RBSCs with lower residual silicon content and higher flexural strength which can be as high as 598 ± 112 MPa.

Keywords: A. Tape casting; C. Strength; D. SiC

Introduction

Reaction bonded silicon carbide have been widely studied and commercially used for many years, owing to its advantages such as lower fabrication temperature and easier to form components with complex shape, near-net size and high density, as compared with other sintering method. Conventional method for obtaining RBSC involves infiltrating the molten silicon or silicon alloys into porous preforms containing carbon [1,2]. As for the preparation of porous green bodies, several methods have been reported, including slip casing, pressure casting and dry pressing [2–4]. In the literatures, tape casting method have also been used to form carbonaceous mixture layer for joining silicon carbide ceramics [5], but it is scarcely used to prepare RBSC ceramics to our knowledge [6].

It is generally recognized that tape casting is a reliable and cost effective method for producing thin, flat ceramic sheets, especially for multilayered composite materials [7–9]. Moreover, green bodies prepared by tape casing always contain evenly distributed and interconnected porosity when the organic components were pyrolyzed [10], which is essential for the infiltration of melt silicon phase.

In the present work, we have fabricated the porous SiC/C preforms with different C/SiC ratio by tape casting method and developed the RBSCs by infiltrating of pure silicon into them. However, it was found that the samples were easily cracked after filtration due to the poor strength of the green bodies. In order to enhance the porous preforms, we infiltrated the porous preforms with phenolic resin solution before silicon infiltration. Results showed that the developed RBSCs samples have lower residual silicon content and great increases in flexural strength were achieved.

Experiment procedure

Commercially available α-SiC powder (FCP-15, d_{50} = 0.4 μm) and carbon black (d_{50} = 0.5 μm) were used as the starting materials. Polyvinyl butyral (aircraft-quality, viscosity: 15–35 s) were used as the binder. Butyl benzyl phthalate (BBP) and polyvinylpyrrolidone (K-30) were added as plasticizer and dispersant, respectively. The solvent was the azeotropic mixture of ethanol and butanone (34/66 by weight).

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Firstly, the carbon blacks were added in the solvent mixture with 4 wt% dispersant. The mixtures were ball milled with silicon carbide media for 4 h. Then the SiC powders were added in followed by ball milling for another 6 h. The weight ratio of C:SiC varied from 0.14 to 0.5. After that, the binder and plasticizer were added into the slurry and ball milled for 48 h to ensure the slurries homogeneity. Finally, the slurries were applied for tape casting on a Procast Precision Tape Casting Equipment (Division of International, Inc., Ringoes, NJ) with the gap height of 0.3 mm, and a carrier film speed of 100 mm/min. After drying at room temperature for 4–6 h, the green tapes were cut into the size of 40 mm × 40 mm and laminated at 140 °C for 30 min in order to ensure good adhesion between the tapes. Then, the laminated green bodies were heat treated at 500 °C in vacuum condition to burn out the binder. Infiltration of phenolic resin solution was carried out with vacuum impregnation method and the solution concentration was 50 wt%. After that, the impregnated specimens were dried at 80 °C for 10 h, followed by setting at 150 °C and then pyrolyzed at 1000 °C.

Finally, the porous preforms with or without the impregnation of phenolic resin were infiltrated with molten Si at 1450 °C in vacuum for 30 min. The sintered bodies were then cut and machined to the sizes of 2 mm × 2.5 mm × 25 mm.

The porosity and pore-size distribution of the green bodies were characterized by mercury intrusion method. The density and apparent porosity of the RBSCs were measured using Archimedean method. The volume fraction of free silicon in the specimens was calculated by comparing the weight loss before and after etching in the mixture of 70 wt%HF–30 wt%HNO_3 for 16 h. The 3-point flexure strength of the sintered specimens was tested on an Instron 5566 testing system with the span and the crosshead speed as 20 mm and 0.5 mm/min, respectively. The microstructures of the porous green preforms and the sintered bodies were examined using a scanning electro microscopy (S-4800, Hitachi).

Results and discussion

Fig. 1 shows the typical dried green tape (Fig. 1a) and the microstructure of the porous green preforms after binder removal at 500 °C (Fig. 1b and c). It can be observed that the obtained green tapes were smooth and there were no obvious defects such as holes or cracks. Moreover, they had good flexibility for free bending and cutting. The microstructure of the heat treated green bodies show that the three phases of silicon carbide, carbon blacks and pores were homogeneously distributed (Fig. 1c).

Fig. 2 shows the typical pore-size distribution of the porous green bodies. It can be seen that, the pore sizes distribution were narrow and the pore sizes ranged from 0.02 μm to 0.2 μm. The porosities for all the preforms were all in the same level around 48 vol.%.

Fig. 3 presents the corresponding microstructure of the polished surfaces of the reaction bonded SiC ceramics. The interconnected gray colored phase is SiC and the white phase is the residual free Si. The two phases of silicon carbide and Si were uniformly distributed in the substrate which means that tape casting is an effective way to obtain RBSCs with homogeneous distribution of the Si phase. Additionally, these microstructures show that there are no pores or unreacted

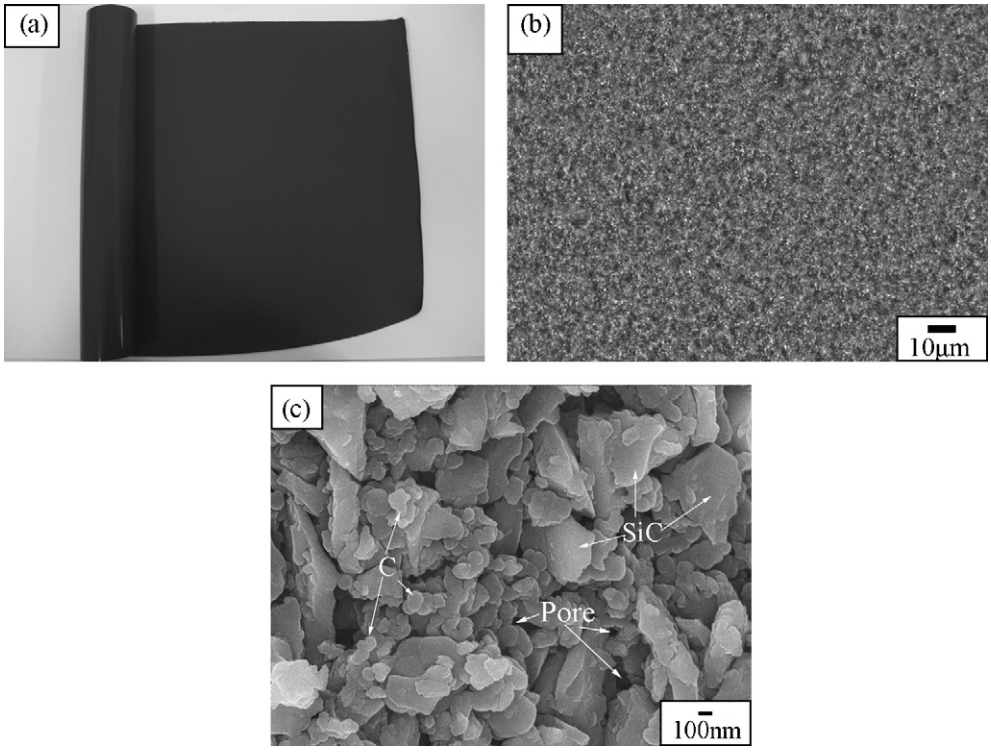

Fig. 1. The typical dried green tape (a) and the microstructure of the porous green preforms after heat treated at 500 °C with amplification of 1000× (b), and 30,000× (c).

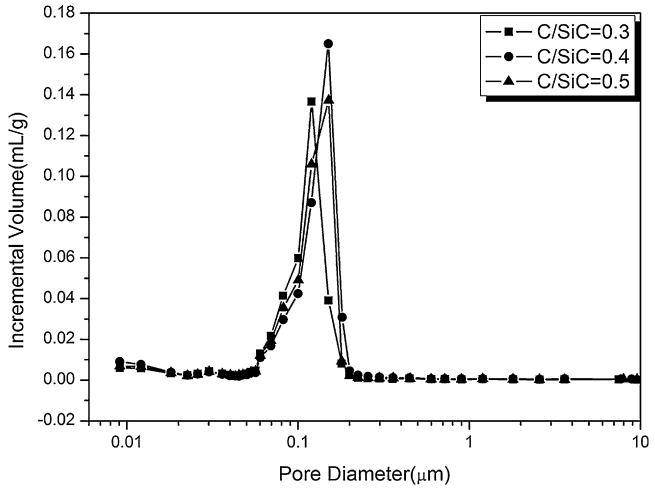

Fig. 2. Pore-size distribution of the porous green bodies with different C/SiC ratios.

carbon in the samples. However, large silicon ''veins'' appeared in the samples with C/SiC ratio as 0.5, which was consequent on the filling of melt silicon into the cracks caused by the volume expansion of newly formed SiC phase during reactive infiltration (Fig. 3d). Previous studies have proved that the process of reactive infiltration of silicon into porous carbon preforms always accompanied with the highly exothermic reaction and volume expansion [4,11].

As the content of carbon black increased, the exotherm reaction would become more significant and the correspondingly thermo stresses resulting from the CTE mismatch between the silicon carbide and the residual silicon would increase. Consequently, cracks were easily to appear in the green preforms with higher carbon content during infiltration if the porous green bodies were not strong enough. The melt silicon would fill in and form silicon ''vein'' in the samples after cooling.

The density and mechanical properties of the RBSCs fabricated from preforms with different C/SiC ratio are shown in Table 1. It is found that the density of RBSCs increased with the increase of the C/SiC ratio. This is for the reason that, with the same porosity, the amount of newly formed silicon carbide would increase with the increasing of carbon content, whereas the space for the residual free Si phase would decrease and the density would decrease accordingly.

The flexural strength of the RBSCs was firstly increased as the C content increased and reached 410 ± 14 MPa when the C/SiC ratio was 0.3 (Table 1). The increasing tendency may be due to the decrease of residual silicon phase as mentioned in the literatures [2,12]. However, the flexural strength of the RBSCs dropped obviously as the C content increased further, which was caused by the more serious thermal stress and the easier formation of Si ''veins'' (Fig. 3d).

In this work, we proposed a simple route to enhance the porous preforms by impregnation of phenolic resin solutions. As shown in Table 1, the residual silicon of the obtained RBSC ceramics decreased with phenolic resin impregnation, while the corresponding density and the bending strength of the sintered bodies increased obviously. Especially for the specimen with C/SiC ratio was 0.3, the flexural strength with impregnation of phenolic resin increased from 410 ± 14 MPa to 598 ± 112 MPa. The decreasing of residual silicon is because of the fact that the pyrolytic carbon derived from phenolic resin could not only help to increase the carbon content in the porous green bodies but also decrease the porosity by partially filled-in of the initial pores.

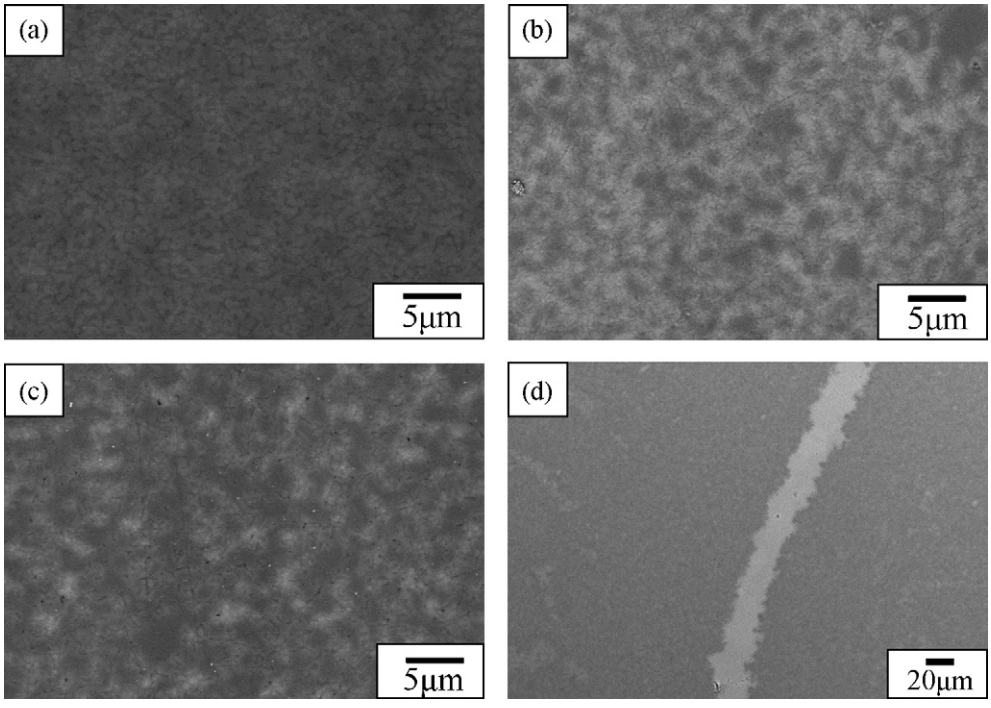

Fig. 3. Microstructure of the polished surfaces of the reaction bonded SiC ceramics with different C/SiC ratio (a) 0.14, (b) 0.3, and (c and d) 0.5.

Table 1
Properties of the RBSCs with or without phenolic impregnation.

Specimen	C/SiC	Density (g/cm^3)	Porosity (vol.%)	Residual Si content (vol.%)	Flexural strength (MPa)
0.14CS	0.14	2.82	0.42	43	335 ± 42
0.2CS	0.2	2.83	0.23	39	367 ± 38
0.3CS	0.3	2.89	0.25	37	410 ± 14
0.4CS	0.4	2.92	0.14	30	352 ± 26
0.5CS	0.5	2.95	0.07	32	225 ± 79
0.14CS + PF	0.14	2.95	1.16	24	430 ± 45
0.2CS + PF	0.2	3.05	0.20	20	549 ± 86
0.3CS + PF	0.3	3.03	0.72	17	598 ± 112

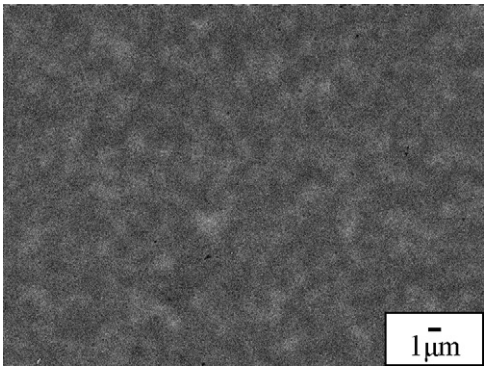

Fig. 4. Microstructure of the polished surfaces of the reaction bonded SiC ceramics with phenolic resin impregnation.

The typical microstructure of the RBSC with phenolic resin impregnation is shown in Fig. 4. It can be seen that the two phases are still well distributed and there are no unreacted carbon or pores.

Conclusion

In summary, RBSCs have been prepared by infiltration of pure Si into the porous preforms fabricated by tape casting method. The green tapes obtained by tape casting method are smooth, uniform and have excellent flexibility for handling. After laminating and de-binding, porous preforms with well-distributed microstructure and narrow pore sizes distribution were obtained. The microstructure of the developed RBSCs show homogeneous and no pore or unreacted carbon was observed. The density of the sintered bodies increase as the increase of the C content while the flexural strength have a peak value when the C/SiC ratio is 0.3, and it reaches 410 ± 14 MPa. Moreover, it was found that infiltration of the porous green body with phenolic resin could help to decrease the residual silicon content and increase the flexural strength of the final RBSCs ceramics. The bending strength of the RBSCs with phenolic impregnation can increase to 598 ± 112 MPa.

Acknowledgements

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